

# PATENT SPECIFICATION

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## (54) CATALYTIC STRUCTURE

(71) We, S.A.E.S. GETTERS S.p.A., an Italian Company, of Via Gallarate 215, 20151 Milan, Italy, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to catalytic (or catalyst) structures, and more particularly to a catalytic structure having an improved heat exchange system.

During catalytic processing, the reactants are normally passed through a bed of porous catalyst particle beads or pellets. In many reactions employing organic materials a fouling carbonaceous deposit appears on the catalyst surface. This fouling may become excessive if the reactants or products remain in contact with the catalyst surface for too long. Fouling results in a change of catalytic activity and also reduces the heat dissipation capacity of the catalyst leading to overheated regions or "hot spots".

It is desirable and usually necessary to control accurately the temperature at which a catalytic reaction takes place. If the temperature differs from that required, the reaction may give only a very low yield of reaction products, thereby rendering the process uneconomic. Alternatively, the reaction may otherwise proceed too far by producing the desired reaction products and then immediately changing these products into other undesired products. Alternatively, the reaction may be completely different from that required.

But when the reaction taking place is exothermic or endothermic, the heat of reaction involved may make it difficult to maintain an accurately controlled reaction temperature.

Thus, in an exothermic reaction, for example, the heat evolved may result in a localized over-heating of the catalyst, thereby causing a "hot spot". Unless this heat is dissipated quickly, the "hot spot" can cause

other, even more exothermic reactions to take place, thereby liberating even more heat with a consequent growth in size of the "hot spot". This growth can lead to a chain reaction thermal instability of the catalytic structure. The structure can be consequently damaged, for instance by sintering of the catalyst from the excessive rise in temperature.

With conventional catalyst, in highly exothermic or endothermic reactions, the reaction tends to respectively heat or cool the catalyst most strongly in the region of greatest reaction. Such a difference in heating or cooling causes a gradual temperature gradient or variations throughout the catalytic structure. With such temperature gradients, only a limited portion of the catalyst is at the optimum reaction temperature. Thus, the rest of the catalyst is not working at greatest efficiency, and its lifetime can be reduced. Therefore, the efficiency for the catalytic reaction is lowered.

A catalyst is usually provided in a composite pellet-like form comprising an active or catalytic portion supported on a carrier. The carrier is generally a poor conductor of heat. Even if the pellet-like support is metallic, there is still poor thermal contact between individual pellets, because the points of contact between the pellets are very small. Temperature control is, therefore, difficult in the conventional form of fixed bed or column type catalysts.

For some catalytic reactions it has been proposed to incorporate pieces of metal or other heat conductive material in mixture with the catalyst to facilitate heat transfer to the surroundings. The reactants may also be diluted with non-reactive gases or vapours as a further means of achieving temperature control. The temperature can also be controlled by using low flow rates or low conversion levels so that the amount of heat generated is low, but this causes the yield per unit time to be low and the process therefore to be more expensive.

Inhibitors to control the rates of reaction

have also been adopted, but these can have the disadvantage of chemically attaching the support of other component of the catalytic device. They can even be responsible for the production of side reactions, which give unwanted products from the reactants.

It is also proposed to control the temperature by supporting the catalyst on a continuous metal support such as a wire mesh or by coating the inside of a hollow tube. In these cases, some compromise has to be made between the variously desired properties. For example, if a thin wire is used as a support to permit placing a large amount of catalyst in a relatively small volume, it becomes difficult to remove excess heat quickly and the structure becomes fragile and delicate. If larger diameter wires are used to increase thermal conduction, the size of the reaction chamber increases and the weight therefore also increases. This increase also causes an increase in the expense of constructing and operating the reaction chamber. If an internally coated tube is used, long tubes must be used to have sufficient contact between the catalyst and the reactants. Thus, a relatively high pressure difference between the ends of the tube is required to cause the reactants to flow through the tube. When the catalyst is exhausted the whole tube must be replaced which is an expensive and lengthy process.

A further disadvantage of known catalytic reaction chambers is that it is difficult to control the flow characteristics in order to ensure maximum catalytic activity but minimum obstruction to the flow of reactants.

Some or all of the aforementioned disadvantages also arise in other related processes, for example, the catalytic purification of gases.

The present invention seeks to overcome some or all of these disadvantages by providing a catalytic structure in which particles of catalyst material are affixed to the surface of a metallic sheet, the sheet thereby conducting heat to or from the catalyst particles as required. At least some of the edges of this metallic sheet are situated thermally adjacent to a heat source or heat sink to thereby heat or cool the metallic sheet as required, whereby the catalyst particles tend to be maintained in thermal equilibrium.

Thus according to one aspect of the present invention a catalyst structure comprises:

- A) a thin sheet having two opposite sides and at least one edge, and being formed of a metal of a given hardness,
- B) a large number of catalyst particles having a hardness greater than said given hardness and being partially embedded in a surface of at least one of the sides of the sheet, and
- C) a mass positioned in good conductive

heat exchange relationship with the sheet through said edge.

According to a second aspect of the present invention a catalyst structure may comprise:

- A) a thin sheet having two opposite sides and being formed of a metal of a given hardness,
- B) a large number of catalyst particles having a hardness greater than said given hardness and being partially embedded in a surface of each of said two opposite sides of said sheet,
- C) said sheet being folded to form a plurality of smaller sheets joined at edges of said smaller sheets to adjacent smaller sheets, and
- D) a mass positioned in good conductive heat exchange relationship with said smaller sheets through at least some of said edges.

According to yet another aspect of the present invention a catalyst structure may comprise:

- A) a thin sheet of metal of a given hardness, said sheet having two opposite sides,
- B) a large number of catalyst particles having a hardness greater than said given hardness, some of which are partially embedded in one of said two opposite sides and some of which are partially embedded in the other of said two opposite sides,

the metal plate having originally a first plurality of pairs of depressions repetitively spaced along one of said two opposite sides, each pair in said first plurality being arranged to facilitate a fold to create a wide edge of a first predetermined type on the other of said two opposite sides of said sheet adjacent the said pair in the first plurality of pairs of depressions, and a second plurality of pairs of depressions repetitively spaced along the said other of said two opposite sides, each pair in said second plurality being arranged to facilitate a fold to create a wide edge of the same or a second predetermined type on said one of said two opposite sides adjacent the said pair in the second plurality of pairs of depressions,

- C) an outer sleeve having a substantially cylindrical inner surface,
- D) an inside support having a substantially cylindrical outer surface, said sheet being folded at the pair of depressions in the first and second plurality of pairs respectively to create the wide edges of the said first and second predetermined types, said wide edges of the said first type being lined up in good thermal contact with said inner surface and said wide edges of the said second type being lined up in good thermal contact with said outer surface, and

E) a fluid piping system in good thermal contact with said outer sleeve for conveying a fluid at a temperature suitable for heat transfer between the fluid and the outer sleeve,

whereby to maintain the catalyst particles in good temperature stability.

Other features of the present invention will appear from the following description and from the claims set out at the end of this specification.

Some embodiments of the present invention also have the advantage that the catalysing structure can be easily removed and changed without extensive changes in the remaining structure of the reaction chamber being used. Furthermore they provide a large amount of catalytically active material in a small volume without impeding the flow of reactants.

Particular embodiments of the present invention, by giving a more uniform temperature within the catalysing structure also allow the use of a higher reaction temperature without incurring the disadvantages of previous catalyzing reaction chambers. Some embodiments will now be described in greater detail, as examples, with reference to the accompanying drawings in which:—

Figure 1 is a cross-sectional view of a metallic sheet with partially embedded catalyst particles for use in the present invention,

Figure 2 is an end view of a reaction chamber utilizing a catalyzing cartridge of the present invention,

Figure 3 is a cross-sectional view along the line A—A' of Figure 2,

Figure 4 is an end view of a reaction chamber utilizing another catalyzing cartridge of the present invention,

Figure 5 is a cross-sectional view along the line B—B' of Figure 4,

Figure 6 is an end view of a reaction chamber utilizing yet another catalyzing cartridge of the present invention,

Figure 7 is an end view of a reaction chamber utilizing another catalyzing cartridge of the present invention,

Figure 8 is an end view of a reaction chamber utilizing a further catalyzing cartridge of the present invention,

Figure 9 is a cross-sectional view along the line C—C' of Figure 8,

Figure 10 is an end view of a reaction chamber utilizing yet a further catalyzing cartridge of the present invention,

Figure 11 is a partly cross-sectional view along the line D—D' of Figure 10,

Figure 12 is an end view of a reaction chamber utilizing another catalyzing cartridge of the present invention,

Figure 13 is a partially cross-sectional perspective view of a point of contact between a folded sheet and an outer sleeve, showing

a fluid-operated heat exchanger for use with the outer sleeve,

Figure 14 is a perspective view of another catalyzing cartridge,

Figures 15 and 16 show details of a thermal contact between a sheet and a sleeve,

Figure 17 shows a transverse cross section through a preferred catalyzing cartridge of the invention,

Figures 18, 19 and 20 illustrate a preferred method of folding a sheet used in the embodiment of Figure 17, and

Figure 21 illustrates, in block form, a preferred use of a catalyst structure of the invention.

Figure 1 is a cross-sectional view of a metallic sheet 1 having catalyst particles 2 and 3 affixed to opposite sides thereof. The construction of such a sheet is fully explained in U.S. Patent No. 3,746,658 (Application Serial No. 33,695, filed May 1st, 1970) relating to an invention of Paolo della Porta, Tiziano Giorgi, Bruno Kindl and Mario Zucchinelli. In the preferred embodiment, a large number of catalyst particles 2 and 3 of a harder material than sheet 1 are affixed to the metallic sheet by a rolling process which partially embeds the particles in the sheet. Although, for the present invention, the metallic sheet preferably has catalyst particles affixed to both sides thereof, a sheet can be prepared and used with catalyst particles on only one side.

These catalyst particles generally pass a U.S. standard screen of 10 mesh per inch and preferably pass a screen of 100 mesh per inch. They should be retained on a screen of 600 mesh per inch. The catalysts which might be used include, but are not limited to, alumina, zirconia, ferric oxide, ferrous oxide, iron oxide, nickel oxide, zinc oxide, potassium chromate, alumina chloride, tungsten sulphide, silver fluoride, cupric chloride, Ag, Zr, Pt, Au, Ir, Ni, Co, Fe, Os, Mn, Mo, Rh, etc.

Figures 2 and 3 disclose a basic embodiment of the present invention, wherein Figure 3 is a cross-sectional view taken along line A—A' in Figure 2. A catalytic structure 10 is shown arranged within a cylindrical sleeve or housing 11 to form a reaction chamber. A metallic sheet having catalyst particles affixed to both sides has been folded into a star-shape form 12 about a central axis. The outside diameter of the star-shaped cartridge thus formed is preferably made slightly larger than the internal diameter of the catalytic chamber housing such that upon placing the cartridge within said housing the extremities of the catalytic structures exert a force against the housing. The cartridge is held firmly but removably in position and is in good thermal contact with the housing.

Any metal may be used as the substrate

for the catalytic material provided that its hardness is less than that of the said catalytic material. However, the substrate should not chemically react with the reactants or with the catalysis products. The substrate should also have a thermal conductivity as high as possible. It should also have good elastic properties if the good thermal contact is required to be provided by the exertion of forces. One preferred substrate metal is aluminium, while bronze, stainless steel, nichrome and copper can also be used successfully.

Star-shaped form 12 is formed of a larger sheet folded to form a number of smaller metallic sheets such as sheet 13 joined to each other at the edges of the sheets, such as edges 14 and 15. At least some of the edges of the sheet, in this embodiment edge 14, are forced against the inside wall 16 of housing 11 for good thermal contact. Thus, the housing 11 can be used as or for connection to a heat source or sink. Flanges 17 and 18 are provided to connect the catalytic reaction chamber to a reaction product input and a reacted product output system (not shown). An external heating or cooling apparatus (not shown) can be placed around housing 11.

Reactants enter the reaction chamber by an entrance port 19, flow through the chamber where catalytic reactions take place, and then reaction products and uncatalyzed reactants leave the chamber by an exit port 20. The flanges can also be used to attach several reaction chambers in series or several cartridges can be placed in series within one housing.

Figures 4 and 5 show an alternate preferred embodiment of the invention. Figure 5 is a cross-sectional view taken along line B—B'. A catalytic reaction chamber 30 has been provided which is identical to chamber structure 10, 11 except for the addition of a centrally placed supporting structure 24 which can also act as a second heat source or sink. The internal edges 15 of catalytic structures such as sheet 13 are in good thermal contact with support structure 24 which is also provided with end flanges 25 and 26. The internal edge 15 of each sheet can be held in intimate contact with support 24 by means of mechanical pressure or alternatively by soldering, welding or other convenient means. Elements numbered the same may be constructed in similar manner to those in Figures 2 and 3.

Figure 6 illustrates a catalytic reaction chamber 40 in which sleeve 11 and supporting structure 24 hold a group of individually formed metallic sheets such as sheet 41, each coated with catalyst particles. The edges 42 and 43 of sheet 41 are respectively thermally adjacent to structure 24 and sleeve 11 of the chamber, which serve as heat-source or heat-sink elements.

Figure 7 illustrates another catalytic reaction chamber 50 including a sleeve 11 and a supporting structure 24. A first catalyzing cartridge composed of sheets, such as sheet 52, has the edges of its sheets, such as edge 53, located thermally adjacent to the sleeve. A second catalyzing cartridge composed of sheets, such as sheet 54, has the edges of its sheets, such as edge 55, located thermally adjacent to the supporting structure 24.

Figures 8 and 9 illustrate an additional catalytic reaction chamber 60 according to the present invention. Figure 9 is a cross-sectional view taken along section line C—C' in Figure 8. Between the supporting structure 24 and the sleeve 11 is placed a catalyzing cartridge folded as a spirated star from a group of spiroid metallic sheets such as sheet 61. Sheet 61 has its two edges 62 and 63, respectively, thermally adjacent to sleeve 11 and supporting structure 24. Preferably, of course, the edges of adjacent spiroid sheets are joined to each other, having been formed by folding a larger sheet.

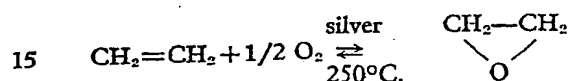
Figures 10 and 11 disclose a catalytic structure 70 according to another embodiment of the invention. Figure 11 is a part cross-sectional view taken through line D—D' in Figure 10. The structure 70 is formed by folding a larger sheet into a star-shaped structure formed of sheets such as sheet 71 having edges 72 and 73. The star-shaped structure is retained by metallic rings 74, which are in thermal contact with the edge 73 of each sheet such as sheet 71. A housing 75 is provided in thermal contact with the rings, so that heat is freely conducted between the catalytic structure and the housing via the sheets and the rings. The rings 74 are provided with a slit or gap 76 so that the catalytic cartridges can be easily inserted into the housing.

Figure 12 shows a catalytic reaction chamber 80 in which the catalytic cartridge comprises an array of involute-shaped catalyzing structures 81 having edges 82 and 83. The curvature of these structures is such that the distance  $d'$ ,  $d''$  between any two structures measured tangentially to a base circle not shown remains constant, thus allowing the dynamic flow characteristics of the reacting gases to be controlled.

#### Example

A catalyzing device according to Figures 2 and 3 is constructed in which the housing 11 is made from stainless steel. A metallic plate 13 is aluminium, of thickness 0.2 mm, is coated by partially embedding thereon by known techniques a finely divided catalytic material, which is a mixture of 70% silver of hardness greater than that of the aluminium substrate, and of 30% alumina which passes through a U.S. standard screen of 100 mesh per inch and is retained on a

U.S. standard screen of 6000 mesh per inch. The catalytic material is partially embedded in a density of 50 mg/cm<sup>2</sup> on both sides of the aluminium strip. The strip is folded into a star-shaped form having an internal diameter of 1 cm, and an external diameter of 5 cm, and is inserted in the housing. The number of folds is such that the cartridge contains 400 gms of silver/liter volume occupied. The outer housing 11 is heated by an oil bath to 250°C, and a mixture of oxygen and ethylene is passed through the catalytic reaction chamber, causing a reaction of the following nature to take place:



A mixture of unreacted reactants, undesired products and ethylene oxide leave the reaction chamber to be separated by means which which form no part of this invention.

Figure 13 is a cutaway view of the point of contact between the sleeve 11 and the plate 13 as illustrated generally in Figures 2—5. A groove 90 is preferably cut into the inside wall 16 of the sleeve or housing to receive the folded edge 14 of sheet 13 and the contiguous sheet. A greater area of thermal contact is provided between the sheet edge and the sleeve by the use of groove 90 than would be provided by contact between the sheet edge and a relatively smooth inside wall 16. In addition, a heat transfer means 91 can be placed in thermal contact with sleeve 11 for greater heat exchange efficiency. Tubes or pipes, such as tube 92, can be used as means for conveying a fluid having a temperature suitable for adjusting the temperature of the sleeve 11 in the desired sense. The fluid can be either heated or cooled, depending upon whether the catalytic reaction is endothermic or exothermic.

Figure 14 is an additional embodiment of the invention in which the star-shaped catalytic cartridge is welded to the inside of a sleeve cylinder 100 having a gap in a split section 102. The welding is carried out along lines such as (hidden) line 104 on the inside of the cylinder. While welding is technically feasible in any instance, the use of the split cylinder makes it practical, since the cylinder can be slightly compressed to insert it in a more permanent sleeve or housing.

Figure 15 shows, in closer detail, the attachment of a sheet 106 to a sleeve 107 by a line of welding or brazing 108. Of course, the contact could be made, as shown in Figure 16, by mere contact between sheet 106 and a smooth sleeve 107, but the resulting area of contact is small, and this is not the most preferred embodiment of the invention.

Figures 17—20 illustrate a convenient way to form a cartridge which is generally of star-shape, as illustrated for example in Figure 8, or in Figure 12.

In Figure 17 is shown a partial cross-sectional view of several spiroid sheets 110, 111 and 112 arranged in a star-shaped cartridge between sleeve 11 and inner support 24. Each sheet is coated with catalyst and has wide edges, for example edges 113 and 114, situated thermally adjacent respectively to support 24 and sleeve 11.

Figure 18 illustrates, in cross-section, how a sheet, prepared as in Figure 19, can be folded to the form used in the embodiment of Figure 17.

Figure 20 is a cross-sectional view of the dies of a step-and-repeat stamping mill, showing a die 120 with projections 121 and a die 122 with projections 123 for acting on a catalyst-coated sheet 125. Projections 121 create a pair of depressions in the sheet which facilitates forming the folds needed adjacent to the inside support 24, and projections 123 create a pair of depressions for the outside fold adjacent to sleeve 11. Rollers bearing suitable projections could be used in place of a step-and-repeat stamping mill. The pairs of depressions are created at repetitive spaced intervals on the respective sides of the sheet. Such spaced intervals and the resulting folds are clearly illustrated in Figure 17.

Thus, Figure 17 illustrates a sheet of metal folded into several smaller sheets 110, 111 and 112 and others not illustrated. A series of pairs of depressions 130 and 131 are provided on opposite sides of the sheet, as at points 121 and 123 on sheet 125 (Figure 20). The pairs of depressions on one side of the sheet may differ somewhat from those on the other side (see Figure 20) and serve to facilitate folds of first and second types. While these types may differ only in the direction they face, they preferably differ also in construction. While both types of fold are wide to allow good thermal contact, the folds of the first type are wider to take advantage of the generally greater space along the inside surface of sleeve 11 than is available to folds of the second type along the outside surface of inside support 24. A plurality of pairs of depressions of both type are repetitively spaced on respectively alternate sides of the sheet as generally indicated in Figure 17 to allow proper dimensioning of the folded sheet.

Figure 21 illustrates the preferred functioning of the catalytic structure disclosed herein. A first reactant fluid and a second reactant fluid enter the catalytic structure, react in the presence of the catalyst particles, and are expelled as a reaction product. Heat is added or removed as necessary to maintain temperature stability.

## WHAT WE CLAIM IS:—

1. A catalyst structure comprising:
    - A) a thin sheet having two opposite sides and at least one edge, and being formed of a metal of a given hardness,
    - B) a large number of catalyst particles having a hardness greater than said given hardness and being partially embedded in a surface of at least one of the sides of the sheet, and
    - C) a mass positioned in good conductive heat exchange relationship with the sheet through said edge.
  2. A catalyst structure comprising:
    - A) a thin sheet having two opposite sides and being formed of a metal of a given hardness,
    - B) a large number of catalyst particles having a hardness greater than said given hardness and being partially embedded in a surface of each of said two opposite sides of said sheet.
    - C) said sheet being folded to form a plurality of smaller sheets joined at edges of said smaller sheets to adjacent smaller sheets, and
    - D) a mass positioned in good conductive heat exchange relationship with said smaller sheets through at least some of said edges.
  3. A catalyst structure according to claim 2 wherein said mass further comprises:
    - A) an outer sleeve having a substantially cylindrical inner surface, and
    - B) an inside support having a substantially cylindrical outer surface, said folded sheet being arranged between said sleeve and said support with each of said smaller sheets one edge positioned in good heat exchange relationship with said sleeve and another edge positioned in good heat exchange relationship with said support.
  4. A catalyst structure according to claim 3 wherein said sleeve is provided with a gap whereby the diameter of the sleeve can be slightly reduced when the sleeve is radially compressed.
  5. A catalytic structure according to claim 3 or 4 wherein said substantially cylindrical inside surface is grooved to receive the edges of said smaller sheets in close thermal contact.
  6. A catalytic structure according to claim 2 wherein said mass further comprises:
    - A) an outer sleeve having a substantially cylindrical inner surface, and
    - B) means for conveying a fluid of a suitable temperature for exchanging heat with said outer sleeve,
  - 60 said folded sheet being arranged in thermal contact with said outer sleeve for enabling conductive exchange of heat therewith.
  7. A catalyst structure according to claim 2 wherein said mass comprises a plurality of rings thermally adjacent to at least some of said edges, said rings being split to allow the diameter of said rings to be slightly reduced when the rings are radially compressed.
  8. A catalyst structure according to claim 2 wherein said mass further comprises:
    - A) an outer sleeve having a substantially cylindrical inner surface, and
    - B) an inside support having a substantially cylindrical outer surface, and further comprising:
      - C) an additional such folded sheet, one of the folded sheets having at least some of its edges in contact with said inner surface and the other of the folded sheets having at least some of its edges in contact with said outer surface.
  9. A catalyst structure comprising:
    - A) a thin sheet of metal of a given hardness, said sheet having two opposite sides,
    - B) a large number of catalyst particles having a hardness greater than said given hardness, some of which are partially embedded in one of said two opposite sides and some of which are partially embedded in the other of said two opposite sides,
  - the metal sheet having originally a first plurality of pairs of depressions repetitively spaced along one of said two opposite sides, each pair in said first plurality being arranged to facilitate a fold to create a wide edge of a first predetermined type on the other of said two opposite sides of said sheet adjacent the said pair in the first plurality of pairs of depressions, and a second plurality of pairs of depressions repetitively spaced along the said other of said two opposite sides, each pair of said second plurality being arranged to facilitate a fold to create a wide edge of the same or a second predetermined type on said one of said two opposite sides adjacent the said pair in the second plurality of pairs of depressions,
  - C) an outer sleeve having a substantially cylindrical inner surface,
  - D) an inside support having a substantially cylindrical outer surface,
- said sheet being folded at the pairs of depressions in the first and second plurality of pairs respectively to create the wide edges of the said first and second predetermined types, said wide edges of the said first type being lined up in good thermal contact with said inner surface and said wide edges of the said second type being lined up in good thermal contact with said outer surface, and,
- E) a fluid piping system in good thermal contact with said outer sleeve for conveying a fluid at a temperature suitable for heat transfer between the fluid and the outer sleeve,

whereby to maintain the catalyst particles in good temperature stability.

10. A catalyst structure according to any preceding claim, substantially as herein-  
5 before described with reference to and as illustrated in the accompanying drawings.

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FIG.1

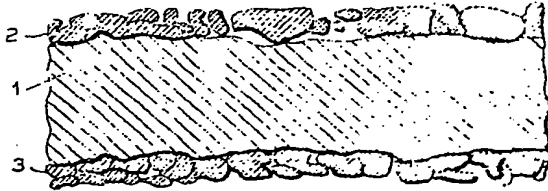


FIG.2

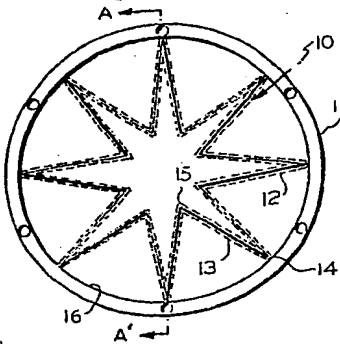


FIG.4

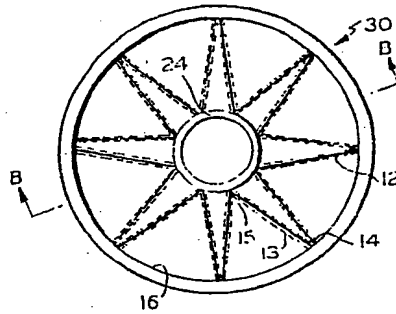


FIG.3

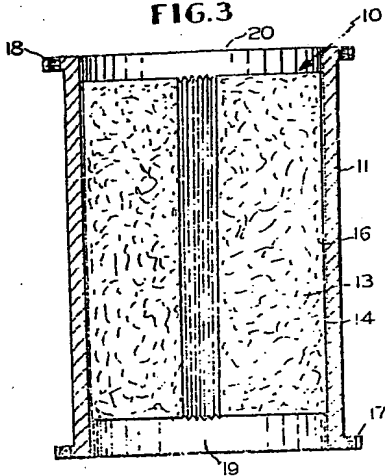


FIG.5

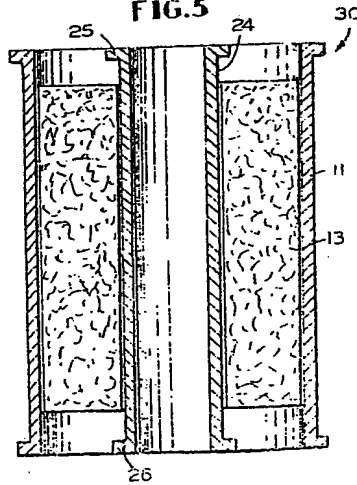




FIG. 6

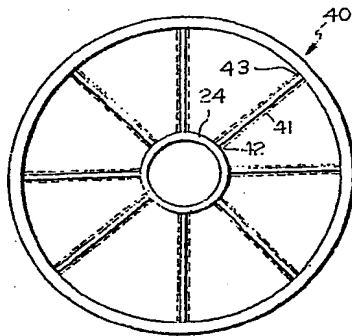


FIG. 7

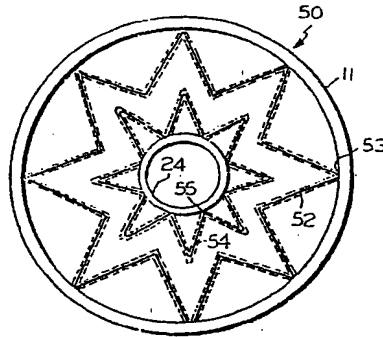


FIG. 8

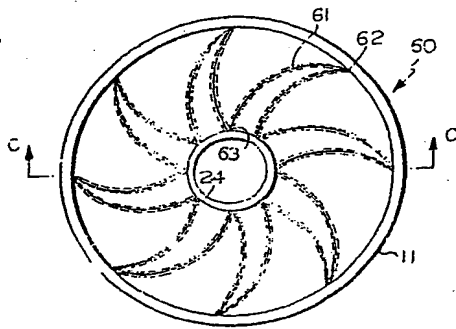
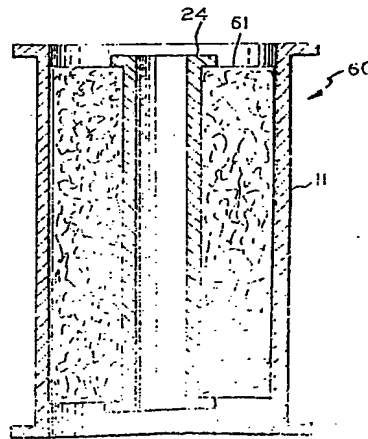
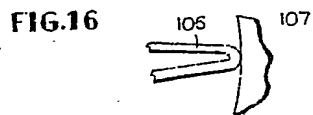
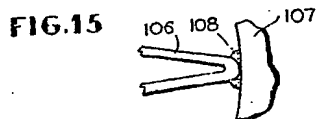
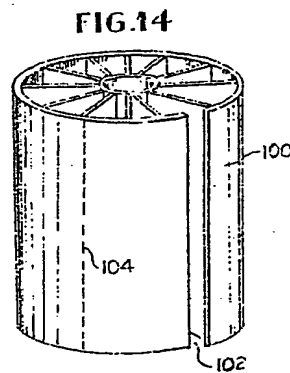
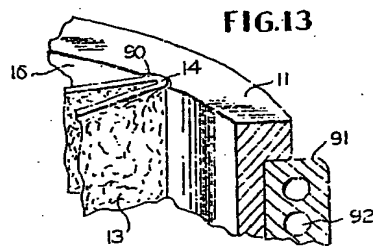
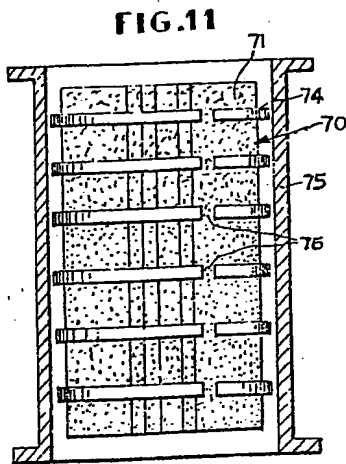
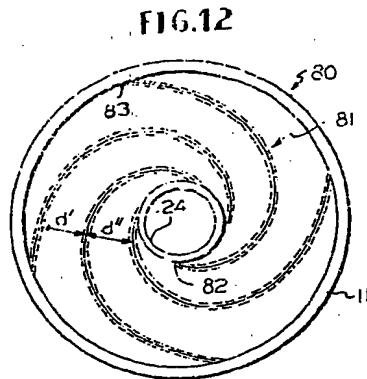
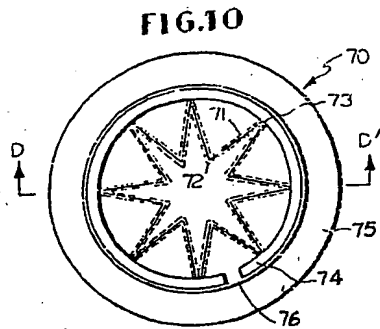


FIG. 9





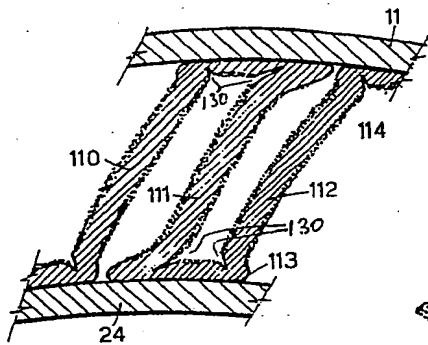


Fig. 17

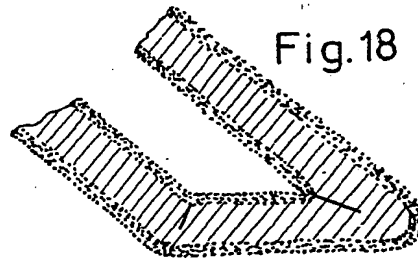


Fig. 18

Fig. 19



Fig. 20

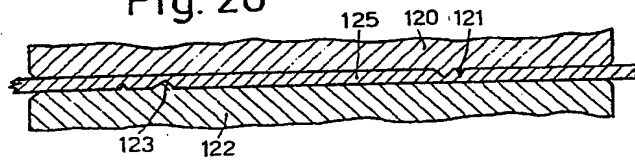


Fig. 21

